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Mobilization of aviation gasoline from a residual source

David W. Ostendorf, Don H. Kampbell, John T. Wilson, John H. Sammons

ABSTRACT: A simple one-dimensional model describes the mobilization of 90 m^3 of residual aviation gasoline from an 80-m diameter, 0.306-m thick contaminated soil mass at the U. S. Coast Guard Air Station in Traverse City, Mich. Originally deposited under a paved ground surface in December 1969, the bound product is thought to be entrained as a separate phase by episodic incursions of the water table in the assumed absence of dissolution, biodegradation, and volatilization. The coupled transport processes of storage, advection, and water table flushing govern the gasoline concentration, subject to a presumed linear sorption relation between mobilized and stationary residual phases. A retardation factor of 13.1 calibrates soil-core data taken at the site from August 1987 to December 1988, with a standard deviation of 26%, indicating a reasonable degree of accuracy. The present position of the leading edge of contamination endorses the calibrated retardation factor value as well. *Res. J. Water Pollut. Control Fed.*, **61**, 1684 (1989).

KEYWORDS: gasoline, groundwater, hazardous wastes, model, soil, spill, transport.

A simple model is derived describing the initial configuration and subsequent mobilization of residual aviation gasoline from a contaminated soil mass at the U. S. Coast Guard Air Station in Traverse City, Mich. The effort is part of a wider investigation by the Robert S. Kerr Environmental Research Laboratory of the U. S. Environmental Protection Agency (EPA) in cooperation with the U. S. Coast Guard into a shallow unconfined aquifer petroleum hydrocarbon plume downgradient of the station.

The plume underlies private residences and has contaminated a number of domestic water wells[1] beginning in 1979 at households some 1000-m downgradient of the suspected source of contamination. The pollution is thought to be due to an accidental release of aviation gasoline in the autumn of 1969. A significant fraction of the spilled hydrocarbons remains bound to the sand and gravel aquifer material as of 1988. This residual fuel acts essentially as a continuous source of organic contamination, in contrast to the instantaneous source behavior usually exhibited by accidental releases of miscible pollutants.

The migration of an immiscible light hydrocarbon spill through the subsurface in its full complexity is a transient nonuniform interaction of liquid product, contaminated water, and the air and vapor phases of both liquids; the interaction is affected by capillarity, differential density, and biological and abiotic transformation.[2-4] Various components of the overall phenomenon have been included in studies with different degrees of sophistication and mathematical detail. Holzer and Hochmuth[5] and Sunada[6] describe the unsteady lateral spread of immiscible light oil over an uncontaminated aquifer using a freshwater-lens analytical solution and a finite-element modeling technique, respectively. Osbourne and Sykes[7] and Lin[8] included capillary effects in two-dimensional transient finite-element models of immiscible fluids in the saturated zone, while Faust[9] and Kuppusamy *et al.*[10] include air as a third immiscible fluid in other finite-difference and finite-element models of unsaturated zone pollution. Separate investigations of solubility, volatility, and biodegradation of organic compound contamination of the subsurface environment in models with varying levels of complexity have been conducted.[11-13] Others[14,15] have proposed sophisticated one-dimensional finite-difference models of more complete versions of the transport problem, including vapor phases and, in the latter analysis, biodegradation of an immobile gasoline constituent.

The conventional concept of dissolved and gaseous hydrocarbon pollution emanating from an immobilized residual source has been brought into question by the Traverse City study, which features gasoline component concentrations well beyond water solubility limits in solid cores far downgradient of the original source area. Pockets of immiscible product have turned up periodically in downgradient groundwater monitoring wells and a model of the dissolved plume constituents[16] implies a moving source term as well, lending additional credence to the notion that separate phases of gasoline components are flowing along the water table some 20 years after the spill. A simple one-dimensional model of the apparent mobilization process, reflects the relatively homogeneous nature of the site, the necessarily sparse spatial and temporal resolution of solid core sampling data, and the depth integrated laboratory analysis of the core material. This source degradation model complements the completed investigations of dissolved downgradient pollution,[16] biodegradation potential,[17] sorptive characteristics,[18] and pump and treat remediation[19] of the Traverse City contaminant plume. The source specification of this present work will be used in ongoing studies of biorestoration, volatile emissions, and mobilized hydrocarbon plume trajectory.

Initial Contaminated Soil Configuration

The suspected source of contamination is a discharge of aviation gasoline through the failed flange of a fuel line.

The flange failure was noticed in December 1969, after the overnight (8 hours) loss of about 9.47 m^3 (2500 gal) of product.[19] This maximum discharge estimate ($Q = 3.29 \times 10^{-4}$ m^3/s) was used to compute the radius (r_j) of aviation gasoline falling through the unsaturated zone in accordance with

$$r_j = \left(\frac{Q\nu}{\pi k g}\right)^{0.5} \tag{1a}$$

$$r_j = 0.382 \text{ m} \tag{1b}$$

Thus, the relatively high soil permeability[1] ($k = 4.75 \times 10^{-11}$ m^2) and low fuel viscosity[20] ($\nu = 6.50 \times 10^{-7}$ m^2/s) yield a thin saturated jet of gasoline descending downward under gravitational acceleration (g) towards the aquifer; in contrast to the more usual case of a broad slow descent of contamination through the unsaturated zone of less permeable material.[2] The actual total release volume is likely to be much higher than 9.47 m^3 (2500 gal) since the fuel line was pressurized continuously before the complete rupture of the flange, which is located 1.3 m below the ground surface as indicated in Figure 1.

The aviation gasoline density ($\rho_G = 7.07 \times 10^2$ kg/m^3) ensures a radial spread of contamination as the jet descends on the capillary fringe of the denser underlying water of the saturated zone. In the assumed absence of soil heterogeneities (an assumption supported by the uniformity of core sample material), one can expect a disk of pollution lying on top of the aqueous capillary fringe, some 4.2 m below the ground surface, with its vertical axis through the location of the failed flange. The vertical extent of contamination is dictated by the capillary rise due to a tensile head (ψ) exerted by the soil grains on the spreading hydrocarbon, as quantified by LaPlace's equation.

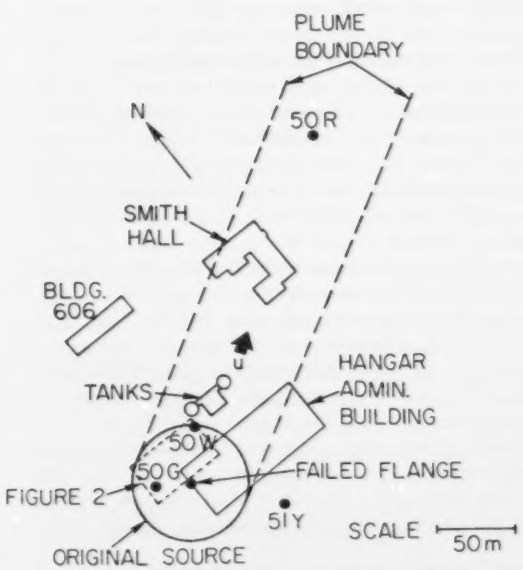

Figure 1—Traverse City, Mich., site area with soil core locations.

$$\psi = \frac{2\sigma}{\rho_G g r} \tag{2}$$

The surface tension[21] ($\sigma = 2.10 \times 10^{-2}$ kg/s^2) across the gasoline-air interface pulls on the liquid with a stress that varies inversely with the radius (r) of the interface. Indeed, it is quite appropriate to think of ψ as the height to which the gasoline will rise in a pore of radius (r) due to capillary attraction of the fluid to the pore walls. Equation 2 includes an assumption of a small contact angle at the grain surface.[21]

The capillary rise extends to a maximum height (H) determined by the smallest continuous pore. At this limiting radius (r_R), the gasoline breaks up into discontinuous pendular masses that are immobile in the unsaturated zone.[3] Microscopic inspection of solid core samples pulled from the site area suggests that r_R is about 2.0×10^{-5} m, and this value may be used with Equation 2 to estimate the thickness of the contaminated soil mass.

$$H = \frac{2\sigma}{\rho_G g r_R} \tag{3a}$$

$$H = 0.306 \text{ m} \tag{3b}$$

The computed thickness compares favorably with intervals of contamination observed in the soil cores obtained from the Traverse City site.

In the absence of a definitive estimate of the accidental release volume, one must assume a value for the initial total residual gasoline content (θ_I) attained on immobilization of the entire spill.

$$\theta = \frac{\text{gasoline volume}}{\text{total volume}} \tag{4a}$$

$$\theta_I = 0.059 \tag{4b}$$

The estimate is based on data[22] for a medium sand at field moisture content and is consistent with the laboratory observations[23] for uniform glass beads as well. The assumed release volume (V) is thus originally configured as a disk of height (H) and a diameter (D) of 80 m, with the latter value set equal to the observed width of the contaminant plume, as shown in Figure 1.

$$V = \frac{\pi D^2 H \theta_I}{4} \tag{5a}$$

$$V = 90.7 \text{ m}^3 \tag{5b}$$

Accordingly, a release of about 90.8 m^3 (24 000 gal) of aviation gasoline to the subsurface at Traverse City can be estimated. This corresponds to an initial total residual source concentration (T_I) equal to

$$T = \frac{\text{average mass}}{\text{wet-soil mass}} \tag{6a}$$

$$T_I = \frac{\theta_I \rho_G}{\rho_B} \tag{6b}$$

$$T_I = 2.12 \times 10^{-2} \tag{6c}$$

Equation 6c reflects an estimated wet-soil bulk density (ρ_B) of 1970 kg/m^3, based on a porosity (n) of 0.367 and

an 80% water saturation. The porosity value is obtained by weighing known solid core sample volumes with subsequent gravimetric analysis of the moisture.

Biodegradation, dissolution, volatilization, and separate-phase mobilization represent possible loss mechanisms for the initial 1969 disk of total residual product mass. The first process is excluded from further consideration since the area overlying the spill was paved for the duration of the contamination, reducing the influx of oxygen and oxygen-saturated water infiltration to the contaminated soil. Volatile losses to the atmosphere are likewise neglected due to the reduced gaseous diffusivity through the paved surface. Dissolution is also ignored in the determination of aviation gasoline source concentrations since the solubility of the primary constituents of aviation gasoline are quite low. In the latter regard, Table 1 displays the results of a gas chromatography/mass spectrometry (GC/MS) analysis of free product obtained from a monitoring well on the site: note the scarcity of soluble benzene-toluene-xylene (BTX) constituents in the sample. In the assumed absence of biodegradation, volatilization, and dissolution, attention is focused on base flow mobilization during periods of high water table elevation; an appropriate emphasis given the proximity of the bulk of the aviation gasoline to the free surface at Traverse City.

Mobilized Source Degradation

The total residual aviation gasoline is presumed to exist in stationary (S) and mobilized residual (M) fractions, with the latter expressed as mass of gasoline per volume of void space.

$$T = S + \frac{Mn}{\rho_B} \tag{7a}$$

$$S = K_D M \quad \text{(linear sorption)} \tag{7b}$$

Table 1—Weathered aviation gasoline* composition.

Constituent	Mass fraction	Molecular weight, kg/mole	Mole fraction
2,2,4-trimethylpentane	0.204	0.114	0.199
2,3,4-trimethylpentane	0.141	0.114	0.137
2,3,3-trimethylpentane	0.134	0.114	0.130
2,3-dimethylpentane	0.123	0.100	0.137
2,2,5-trimethylhexane	0.087	0.128	0.075
Toluene	0.047	0.092	0.057
2,4-dimethylpentane	0.039	0.100	0.043
2,2,3,4-tetramethylpentane	0.034	0.128	0.029
2,3-dimethylhexane	0.030	0.114	0.029
2,3-dimethylbutane	0.027	0.086	0.035
2,5-dimethylhexane	0.023	0.114	0.022
2,3,5-trimethylhexane	0.010	0.128	0.009
2,2,5,5-tetramethylhexane	0.006	0.142	0.005
Remainder	0.095	0.114 (assumed)	0.093

* Free product from monitoring well M54; product density is 707 kg/m^3.

This study considers the coupled transport of these partitions, which are presumed to be related by linear sorption with an empirically determined distribution coefficient (K_D). The mobilized conservation of mass equation is taken as a simple one-dimensional balance of storage, advection, and water table flushing, subject to a retardation factor (R_D)

$$R_D \frac{\partial M}{\partial T} + \frac{u}{n}\frac{\partial M}{\partial x} + \frac{iM}{nH} = 0 \quad \text{(mobilized)} \tag{8a}$$

$$R_D = 1 + \frac{\rho_B K_D}{n} \tag{8b}$$

$$S = T\left(1 - \frac{1}{R_D}\right) \tag{8c}$$

with time (t) and downgradient distance (x) from the leading edge of contamination. The advection proceeds down the gradient of hydraulic head at the time-averaged specific discharge (u); the vertical flushing is due to the water table fluctuation rate (i). It is important to distinguish the use here of R_D and K_D from the usual application of linear sorption. A separate-phase material held in the solid matrix by capillary forces is considered in equilibrium with an emulsion; conventional sorption describes an instantaneous balance between chemically adsorbed and dissolved material. Equation 8c suggests that the total residual hydrocarbon concentration is essentially equal to the stationary residual gasoline content for the case of strong sorption, which occurs at Traverse City.

The transport equation is in force only during periods of contact between the saturated aquifer base flow and the originally contaminated soil mass, as reflected in reduced time-averaged values for u and i. In the absence of historical records, the water table is presumed to flush one pore volume through the contaminated soil mass during the wet season of the water year over a 2-month period. The specific discharge through the source mass is thus equal to one-sixth of the actual value below the water table at Traverse City and the water table fluctuation rate is set at one pore volume per year.

$$u = \frac{u\,(\text{actual})}{6} \tag{9a}$$

$$u = 2.89 \times 10^{-7}\ \text{m/s} \tag{9b}$$

$$i = \frac{nH}{1\ \text{year}} \tag{9c}$$

$$i = 3.56 \times 10^{-9}\ \text{m/s} \tag{9d}$$

A uniform initial concentration and a pure upgradient ambient flow constrain the mobilized residual hydrocarbon transport equation.

$$M = \frac{S_I}{K_D} \quad (t = 0) \tag{10a}$$

$$M = 0 \quad (x = 0) \tag{10b}$$

Equation 8a may be recast in method of characteristics form by considering the temporal concentration variation (dM/dt) in frames of reference moving at speed dx/dt.

$$\frac{dM}{dt} = -\frac{iM}{R_D n H} \quad (11a)$$

$$\frac{dx}{dt} = \frac{u}{R_D n} \quad (11b)$$

Equation 11a may be easily integrated from 0, S_I/K_D to t, M, resulting in an exponential decay of concentration in the moving reference frames, whose positions are established by integration of Equation 11b. The frame leaving the leading edge of contamination at $t = 0$ will separate clean and contaminated water, so that our solution may be expressed as

$$M = \frac{S_I}{K_D} \exp\left(-\frac{it}{R_D n H}\right) \quad \left(t < \frac{R_D n x}{u}\right) \quad (12a)$$

$$M = 0 \quad \left(t > \frac{R_D n x}{u}\right) \quad (12b)$$

The mobilized concentration is thus maintained at an unsteady, spatially uniform concentration (Equation 12a) until the arrival of the reference frame traveling at the retarded speed $u/R_D n$ from the leading edge of contamination, after which the pollution vanishes (Equation 12b). The stationary residual aviation gasoline concentration follows simply from the mobile-phase solution and the condition (7b) of linear sorption

$$S = S_I \exp\left(-\frac{it}{R_D n H}\right) \quad \left(t < \frac{R_D n x}{u}\right) \quad (13a)$$

$$S = 0 \quad \left(t > \frac{R_D n x}{u}\right) \quad (13b)$$

Equations 13a and 13b describe the degradation of the original stationary residual aviation gasoline by mobilization in a fluctuating water table. The process is mediated by a retardation factor, which was used for model calibration with the soil core data at Traverse City.

Traverse City Soil Core Data

Table 2 lists the total residual aviation gasoline concentration of soil core samples collected from August 1987 to December 1988 at sites specified in Figures 1 and 2. Leach *et al.*[24] described the sampling methodology in detail. Briefly, a soil core 0.50 to 0.75 m long is withdrawn from a sample barrel driven through a hollow-stem auger into undisturbed soil as part of an EPA-developed protocol for biological and geochemical characterization of the subsurface. The core is extruded in sections with minimal head space into sampling jars on-site in a nitrogen-filled glove box equipped with an aseptic paring device to minimize cross-contamination. The core sections with the maximum total residual aviation gasoline concentration

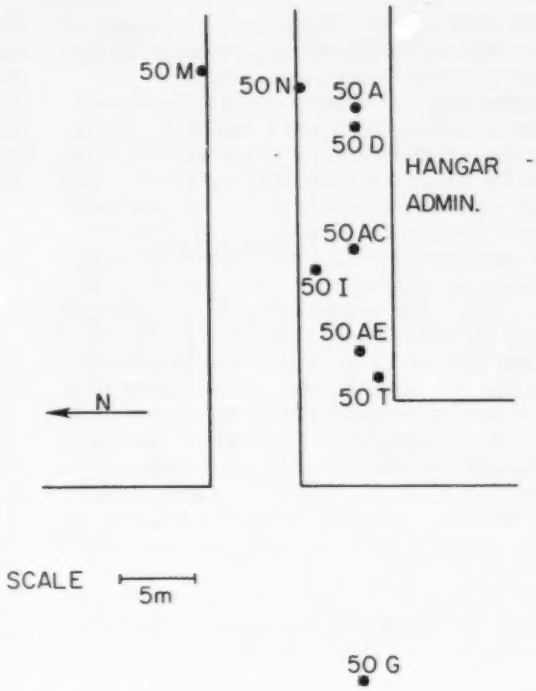

Figure 2—Detail showing soil core locations.

Table 2—Soil core data.

Sample	Date	Elevation, m msl	x, m	S_w, volume water / volume voids	T
51Y	10/88	181.4–184.5	—	ND[a]	BDL[b]
50R-6	5/88	182.4–182.7	220.0	0.93	0.0122
50G	8/87	181.4–184.5	24.0	ND	BDL
50T-3	5/88	182.7–183.0	44.5	1.00	0.0033
50AE-4,5	10/88	182.8–183.0	44.5	1.00	0.0054
50AH-22,23	12/88	182.9–183.1	47.5	0.80	0.0058
50AC-7,8	10/88	182.9–183.1	53.3	ND	0.0068
50I-14	8/87	183.1–183.3	53.4	0.97	0.0065
50A-3	8/87	182.7–183.4	64.4	0.45	0.0056
50N-2,3	3/88	182.3–183.2	65.5	ND	0.0061
50M-2	3/88	182.6–183.2	71.0	ND	0.0025
50W-3	5/88	182.9–183.1	72.5	1.00	0.0048

[a] Not determined.
[b] Below detection limit of 10 mg/kg.

are shown in Table 2; in nearly all cases these samples are bound by soil with much lower levels of contamination, as is implied by Equation 3b. Vandegrift and Kampbell[25] discuss the GC laboratory procedure used to deduce contamination levels in depth-integrated subsamples withdrawn from the sampling jars. Methylene chloride is used to extract the hydrocarbons from the soil sample for subsequent analysis in a wide-bore capillary column.

The data support various qualitative conclusions regarding the distribution of immiscible aviation gasoline in the site area. The 50R data point offers proof of the mobilization phenomenon since it is well beyond the confines of the original source mass, and the 51Y core is free of contamination, as would be expected due to its position outside of the plume. Moisture saturations (S_W) cited in Table 2 suggest that in most cases the maximum contamination lies slightly above the saturated capillary fringe of the aquifer. The distance (x) from the original position of the leading edge of contamination is also included in Table 2; note that the interface of clean and polluted soil has apparently passed core 50G but has not yet arrived at core 50T.

The stationary residual aviation gasoline data are used to calibrate the retardation factor, with the results summarized in Table 3. Equations 6c, 8c, and 13a predict values for residual saturation when R_D is specified. The error (δ) defined by

$$\delta = \frac{S\,(\text{measured}) - S\,(\text{predicted})}{S\,(\text{predicted})} \qquad (14a)$$

$$\bar{\delta} = \frac{1}{j}\,\Sigma\delta \qquad (14b)$$

$$\sigma = \left[\frac{1}{j}\,(\Sigma\delta^2) - \bar{\delta}^2\right]^{0.5} \qquad (14c)$$

is used to specify the parameter and assess model accuracy, with j data points.[26] The error standard deviation (σ) is a measure of the spread of the data about their predicted values, while the mean error ($\bar{\delta}$) is set equal to zero by calibrating the retardation factor with the value

$$R_D = 13.1 \quad (\bar{\delta} = 0) \qquad (15)$$

Table 3—Model calibration results.

Sample	t, s × 10^8	S (observed) S_i	S (predicted) S_i	δ, percent
50T-3	5.81	0.156	0.245	−36
50AE-4,5	5.94	0.255	0.238	7
50AH-22,23	5.99	0.274	0.235	17
50AC-7,8	5.94	0.321	0.238	35
50I-14	5.57	0.307	0.260	18
50A-3	5.57	0.264	0.260	2
50N-2,3	5.76	0.288	0.248	16
50M-2	5.76	0.118	0.248	−52
50W-3	5.81	0.226	0.245	−8

Predictions based on calibrated R_D = 13.1, yielding $\bar{\delta}$ = 0% and σ = 26%.

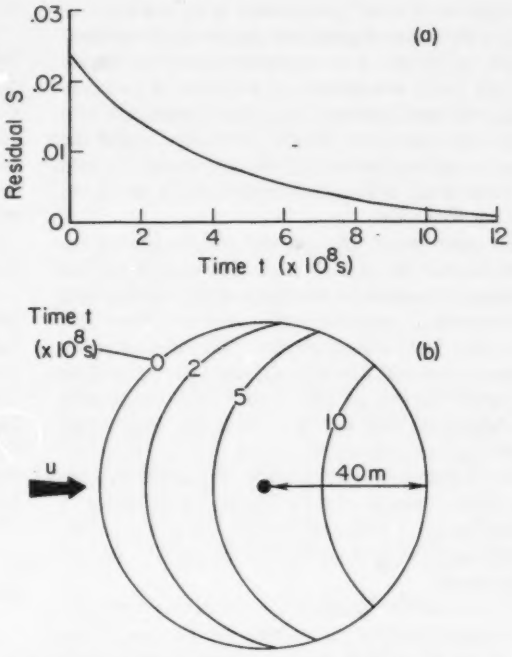

Figure 3—Source strength (a) and plan configuration (b) as functions of time.

Table 3 indicates no systematic error with downgradient distance, and the 26% standard deviation represents reasonable calibration accuracy in light of the sampling errors inherent in the search for a 0.3-m contaminated core interval some 4 m below the ground surface under field conditions.

The position (x_F) of the upgradient boundary of residual aviation gasoline may be estimated from the calibrated R_D value.

$$x_F = \frac{ut}{R_D n} \qquad (16a)$$

$$x_F = 36 \text{ m} \quad (\text{December 1988}) \qquad (16b)$$

Thus the leading edge of contamination presently (December 1988) lies between cores 50G and 50T, in accordance with the field data.

Discussion

The mobilization of residual aviation gasoline from the original contaminated soil mass location has important consequences for Traverse City, in particular, and for nonaqueous-phase liquid transport, in general. Figure 3 displays the strength of the originally contaminated soil mass as a function of time based on the calibrated retardation factor of 13.1. A duration of 1.33×10^9 s (42 years) has been computed for the mobilization of the entire bound product, at which time the stationary residual concentration will fall to a value of 7.8×10^{-4}. This estimate does not reflect biodegradation, dissolution, and volatil-

ization loss which would presumably make small contributions to the source degradation process under ambient conditions at the site. It is important to note that Figure 3 does not imply remediation of any kind; it gradually replaces a stronger stationary mass of contamination with a weaker moving source. Indeed, one would expect the mobilized aviation gasoline to slowly contaminate a much larger mass of soil at lower concentrations in the downgradient environment.

In the latter regard, the episodic entrainment of immiscible product during high water table excursions into the original contaminated soil mass on the site has been postulated with an assumed linear sorption isotherm between mobile and residual phases. This results in pulses of contaminated base flow moving along the free surface at a retarded velocity of 3.61×10^{-7} m/s (u(actual)/$R_D n$). Referring to Figure 1, we accordingly suspect that the mobilized gasoline arrived at the 50R location 4.99×10^8 s (16 years) after the accident, and presently (December 1988) extends roughly 220 m downgradient of the failed flange. A field investigation will further document the trajectory of the mobilized immiscible aviation gasoline plume.

The slowly moving droplets of separate-phase product serve as a concentrated near-surface source term for the traditional mechanisms of dissolved advection and volatile diffusion. In this regard, the dissolved contaminant plume moves about 13 times faster than its immiscible counterpart with anticipated locally strong vapor pressure gradients above the pulses of gasoline, and periodic occurrences of free product in monitoring wells screened near the water table. All these phenomena have been observed to some degree at Traverse City and can be expected wherever immiscible product, initially immobilized immediately above the free surface of an unconfined aquifer, is subsequently subjected to episodes of high water excursion.

Conclusions

A simple model was derived describing the initial configuration and subsequent mobilization of residual aviation gasoline from a contaminated soil mass at the U. S. Coast Guard Air Station in Traverse City. The December 1969 spill volume of about 90 m^3 flows in a thin jet directly to the unconfined aquifer, to spread into a disk of 80 m diameter and 0.306 m height above the capillary fringe. Storage, episodic advection, water table flushing, and linear sorption combine to reduce the original stationary residual concentration in accordance with an exponential decay behind a slowly moving contamination boundary. A rough field calibration of this vertically integrated model yields a retardation factor of 13.1 with an error standard deviation of 26%. The present position of the leading edge of contamination endorses the calibrated retardation factor value as well. The source can be expected to persist at its original location for 40 years at a steadily diminishing strength in the assumed absence of biodegradation, volatilization, and dissolution. The entire spill is expected to be moving slowly through the downgradient environment after this time.

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